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cessed with reduced risk of contamination. Of course, there can be other variations, modifications, and alternatives.

In some embodiments, the autoclave is then heated to crystal growth conditions. In some embodiments, the thermal cycle includes a pre-reaction segment to form mineralizer, polycrystalline gallium nitride, dissolved gallium containing complexes, or the like. In some embodiments, the atmosphere in the autoclave may be modified during the run. For example, excess  $H_2$  formed by reaction of gallium metal with ammonia may be bled off through the gate valve or allowed to diffuse through a heated palladium membrane. The use of a commercially-available heated palladium membrane offers the convenience of continuous hydrogen removal, without interrupting or perturbing the process, with little or no opportunity for contamination of the process. Excess nitrogen formed by decomposition of an azide mineralizer may be bled off through the gate valve. Additional ammonia may be added to replenish the solvent in the high pressure apparatus.

After performing crystal growth for a predetermined period of time, the autoclave is cooled. When the autoclave has cooled to below 100 degrees Celsius, below 75 degrees Celsius, below 50 degrees Celsius, or below 35 degrees Celsius, a valve to the autoclave is opened and the ammonia is removed. In a specific embodiment, gas-phase ammonia is allowed to exit the autoclave and is bubbled through an acidic aqueous solution in order to be chemically trapped. In another embodiment, gas phase ammonia is passed through a flame so as to burn the ammonia, forming  $H_2O$  and  $N_2$ . In a preferred embodiment, illustrated in FIGS. 6A and 6B, the ammonia is trapped for recycling and reuse.

Referring to FIG. 6A, appropriate for a single-ended autoclave, the ammonia may be removed as either a liquid or a gas. To remove the ammonia as a liquid, the inner purge tube (see FIG. 3) is re-inserted into the outer purge or fill tube and the outlet of the inner purge tube connected to the Receiving/Purification tank. Keeping the purge gas exhaust connection (see FIG. 3) closed, a valve in the line to the inner purge tube is opened, allowing liquid ammonia to flow through the inner purge tube in the autoclave into the Receiving/Purification tank, which is otherwise closed. The Receiving/Purification tank may be cooled, for example, by chilled water, and/or the autoclave and transfer line may be heated during the ammonia transfer operation, so as to maintain a higher vapor pressure of ammonia in the autoclave as compared to the vapor pressure in the Receiving/Purification tank. The temperature differential between the autoclave and the Receiving/Purification tank may be held between one and 50 degrees Celsius. In another embodiment, the ammonia is removed as a vapor. The outlet of the autoclave is connected to a condenser above the Receiving/Purification tank and a valve opened. Gas-phase ammonia enters the condenser and condenses into liquid in a heat-exchanger, for example, a chilled-water-cooled coil, at a pressure between about 100 and 250 pounds per square inch. The autoclave and transfer line may be heated to a temperature that is higher than the condenser by between one and 50 degrees Celsius. Residual gases, for example,  $N_2$  and  $H_2$ , may be released by venting to a scrubber and/or a flame.

Referring to FIG. 6B, appropriate for a double-ended autoclave, the ammonia may be removed as a liquid. A port on the bottom end of the autoclave is connected to the Receiving/Purification tank and a valve opened, allowing liquid ammonia to flow into the Receiving/Purification tank, which is otherwise closed. The Receiving/Purification tank may be cooled, for example, by chilled water, and/or the autoclave and transfer line may be heated during the ammonia transfer operation, so as to maintain a higher vapor pressure of ammonia in the autoclave as compared to the vapor pressure in the

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Receiving/Purification tank. The temperature differential between the autoclave and the Receiving/Purification tank may be held between one and 50 degrees Celsius.

For recycling purposes, a purifying agent, for example, sodium metal, may be placed in the receiving/purification tank. The sodium will react with residual oxygen and/or water in the ammonia, restoring a very high degree of purity. After a period of one hour to thirty days, the ammonia may be transferred to a delivery tank. In a preferred embodiment, the transfer is performed through the gas phase, via a condenser, so as to leave the purifying agent in the receiving/purification tank. Liquid ammonia may be delivered from the delivery tank, via a dip tube, to the autoclave for the next crystal growth run. In an alternative embodiment, vapor-phase ammonia may be delivered from the delivery tank to the autoclave for the next crystal growth run.

After removing the ammonia, the autoclave is opened and grown crystals and remaining raw material removed. The crystals may be sliced in a predetermined orientation to form at least one wafer. After slicing, the crystal wafers may be lapped, polished, and chemical-mechanically polished by methods that are known in the art.

In a specific embodiment, any of the above sequence of steps provides a method according to an embodiment of the present invention. In a specific embodiment, the present invention provides a method and resulting crystalline material provided by an autoclave apparatus having means for filling with a solvent at elevated pressure. Other alternatives can also be provided where steps are added, one or more steps are removed, or one or more steps are provided in a different sequence without departing from the scope of the claims herein.

While the above is a full description of the specific embodiments, various modifications, alternative constructions and equivalents may be used. As an example, the present method and system uses a pressure of about 7 atmospheres and greater, but other suitable pressures or specific pressures may exist. In one or more embodiment, the pressure may be slightly higher or lower depending upon the application. Therefore, the above description and illustrations should not be taken as limiting the scope of the present invention which is defined by the appended claims.

What is claimed is:

1. A process for growing a crystalline gallium-containing nitride, the process comprising:
  - providing a high pressure apparatus;
  - providing a capsule comprising gallium-containing feedstock in one zone and at least one seed in another zone; while maintaining an elevated pressure within the capsule, introducing a solvent capable of forming a supercritical fluid into at least the one zone and the other zone;
  - placing the capsule within the high pressure apparatus;
  - processing one or more portions of the gallium-containing feedstock in the supercritical fluid to provide a supercritical solution comprising at least gallium containing species at a first temperature; and
  - growing crystalline gallium-containing nitride material from the supercritical solution on the seed at a second temperature, the second temperature being characterized to cause the gallium containing species to form the crystalline gallium containing nitride material on the seed.
2. The process of claim 1, wherein the solvent capable of forming a supercritical fluid is introduced into the capsule at an elevated pressure of at least about seven (7) atmospheres.